## The role of $CO_2$ purification and transport networks in carbon capture and storage cost reduction

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#### Abstract

A number of Carbon Capture and Storage projects (CCS) are under way around the world, but the technology's high capital and operational costs act as a disincentive to large-scale deployment. In the case of both oxycombustion and post-combustion  $CO_2$  capture, the  $CO_2$  compression and purification units  $(CO_2CPU)$  are vital, but costly, process elements needed to bring the raw  $CO_2$  product to a quality that is adequate for transport and storage. Four variants of the CO<sub>2</sub>CPU were modelled in Aspen HYSYS each of which provide different  $CO_2$  product purities at different capital and operating costs. For each unit, a price of  $CO_2$  is calculated by assuming that it is an independent entity in which to invest and the internal rate of return (IRR) must be greater or equal to the minimum rate of return on investment. In this study, we test the hypothesis that, owing to the fact that  $CO_2$  will likely be transported in multi-source networks, not all  $CO_2$  streams will need to be of high purity, and that it may be possible to combine several sources of varying purity to obtain an end-product that is suitable for storage. We find that, when considering study generated costs for an example network in the UK, optimally combining these different sources into one multi-source transport network subject to a minimum  $CO_2$  purity of 96% can reduce the price of captured  $CO_2$  by 17%.

Preprint submitted to International Journal on Greenhouse Gas Control20th November 2016

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## Nomenclature

Symbol/Acronym	Description
ASU	Air Separation Unit
CAPEX	Capital costs
CCS	Carbon Capture and Storage
CPU	Compression and Purification Unit
CPU, CD	Compression and Dehydration CPU
CPU, Dist	Distillation CPU
CPU, HPDF	High Purity Double Flash CPU
EOR	Enhanced Oil Recovery
IRR	Internal Rate of Return
MILP	Mixed-integer linear programming
OPEX	Operational costs
PostC	Post Combustion

#### 1. Introduction

As Carbon Capture and Storage (CCS) is making its way forward to deployment around the world, one of the principal remaining barriers are the costs involved and a lack of economic incentive for investment. This applies to CCS for the power sector and industrial sectors. In fact, 21% of total global anthropogenic greenhouse gas emissions come from industry of which almost 60% stem from iron, steel and cement production [1, 2]. Whilst the power sector has several options for partial or total decarbonisation including improving efficiency, switching fuels, switching to renewable or nuclear power sources, the same options are not equally available to the heavy industry sector. The industrial sector will always emit process  $CO_2$  and therefore their decarbonisation relies solely on CCS [3]. In this study we discuss the effect of  $CO_2$  purity on transport and storage, the origin of  $CO_2$  stream regulations and the prospect of transport networks in Section 1.1, and the cost-purity trade-off that exists in oxy-combustion  $CO_2$  capture in Section 1.2. Thereafter, in Section 2 and 3, we examine and evaluate the cost reduction opportunities for the  $CO_2$  capture system that stem from having a multi-source to sink  $CO_2$  transport system.

#### 1.1. Background to the link between CO<sub>2</sub> purity, transport and storage

The cost per tonne of  $CO_2$  captured is a key consideration when choosing between capture technologies. In the case of both oxy-combustion and membrane processes, there exists the potential for a trade off between the cost of capturing the  $CO_2$  and the purity at which the final  $CO_2$  product is produced. This is in contrast to ab- or adsorption-based post-combustion capture technologies which, by virtue of their high selectivity, inherently produce a high purity stream of  $CO_2$ . This opportunity arises from the fact that the raw exhaust gas exiting from an oxy-combustion boiler is around 80%  $CO_2$  with the balance composed of  $N_2$ ,  $O_2$ , Ar,  $H_2O$  and other trace elements<sup>1</sup>. It is important to recognise that whilst the final  $CO_2$  storage destination imposes a hard constraint on  $CO_2$  purity<sup>2</sup>, it is possible to safely transport  $CO_2$  containing some impurities such as  $N_2$ ,  $O_2$  or Argon. This purity constraint is partly based on economics and partly based on existing legislation requiring  $CO_2$  to "overwhelmingly consist of  $CO_2$ " [4, 5]. It is also important to note

<sup>&</sup>lt;sup>1</sup>SO<sub>x</sub> and NO<sub>x</sub>

<sup>&</sup>lt;sup>2</sup>Minimum  $CO_2$  concentration of 96% for all storage sites

that whilst near-term demonstration plants, e.g., the Boundary Dam plant in Canada, are single-source to single-sink systems, the same is unlikely to be true once multiple CCS plants are in operation, with transportation networks being more cost-effective [6, 7, 8]. It is therefore sensible to evaluate the potential role of this infrastructure in contributing to the cost reduction of CCS.

It is generally agreed that large volumes of  $CO_2$  (over  $5MtCO_2/year$  [2]) are best transported as a supercritical/dense phase fluid [9] and phase changes associated with pressure or temperature changes should be avoided for safe operation of the pipeline. It must further be recognised that the pressure to which the  $CO_2$  stream needs to be compressed to ensure safe operation of the transport element is a strong function of the composition of that stream [10, 2]. This pressure requirement increases in proportion to the fraction of impurities in the stream as they bring the mixture's two phase region above that of pure  $CO_2$ .

Yan et al. [4] studied the effect of a change in inlet stream composition on the pipeline inlet pressure requirement and have shown that an inlet stream of 87 vol% CO<sub>2</sub> requires a 17% increase in inlet pressure compared to an inlet stream of 96 vol%. Wetenhall et al. [11] has shown that a higher content of non-condensable gases in the inlet stream increases the pipeline inner diameter and thus the relative cost per km of pipeline. However, Yan et al. [4] demonstrated that over a short pipeline distance of 30km, there is no change in pipeline cost with product purity and transporting lower purity  $CO_2$  streams is reasonable over these distances. Hence, in a context in which transport networks are considered - with multiple pipelines joining into a trunk line linking to a storage sink - lower purity  $CO_2$  streams can be inserted into the network as long before joining higher purity  $CO_2$  streams and when mixed in the pipeline will reach desired purities.

Other studies, such as Mahgerefteh et al. [12], modeled the likelihood of propagating fractures in pipelines as a function of the content of noncondensable gases in the  $CO_2$  stream. This theoretical study implied that, if a crack occurs in a pipeline containing an oxy-combustion derived  $CO_2$ stream of 88.4% purity, the crack would propagate creating a long running fracture, whereas a pure  $CO_2$  stream in the same pipeline would not cause the crack to propagate. It is important to note that fracture propagation is a strong function of the pipeline external temperature, this phenomenon is ultimately by the fact that the saturation pressure of an impure  $CO_2$  stream is greater than that of a pure  $CO_2$  stream. Hence, de-pressurisation of the pipeline results in rapid phase change (dense phase to vapour phase) and subsequent cooling, which makes the pipeline prone to fracture propagation [12]. However, it is important to note that these conclusions have not been corroborated by experimental data [13].

The design of the transport system is then a function of the final storage sink chosen: saline aquifers <sup>3</sup> or depleted hydrocarbon reservoirs and the potential to use the stream for enhanced oil recovery (EOR). A  $CO_2$  stream suitable for EOR is commonly regarded as needing to be of very high purity, with limits on  $O_2$  concentration at about 100ppm [16]. High levels of oxygen in the  $CO_2$  stream used for EOR risks causing overheating at the injection point, oxidation and biological growth in the reservoir [17]. However, a number of studies have pointed out that there is a lack of fundamental research and industrial experience with anthropogenic  $CO_2$ . The actual level above which a 'high level' of oxygen is reached and associated risks become plausible remains highly uncertain [17, 18]. This is linked to that fact that most of the regulations and common practices for  $CO_2$ -EOR are based on North-American experience [2] and a large amount of the  $CO_2$  used for EOR in North America came from natural sources of  $CO_2$  at naturally high purity. A typical example of this is the Sheep Mountain  $CO_2$  reservoir in the US that contains a  $CO_2$  stream of 97 vol%  $CO_2$ , 0.6 vol%  $N_2$ , 2.4 vol%  $CH_4$  and trace amounts of water. This stream of natural  $CO_2$  is typically delivered to the oil producers at a pressure of 97 bar and at a temperature below  $24^{\circ}$ C [19]. Given the uncertainty on the risk of certain levels of impurities on  $CO_2$ -EOR, it would be beneficial for the process to relax the strict requirements for  $CO_2$ purity and thus reduce the gap in profitability.

The presence of impurities in  $CO_2$  does also have an impact on storage. The IEA GHG R&D Programme [20] studied the reduction in storage capacity as a result of higher fractions of non-condensables in the  $CO_2$  stream. This study showed that in a saline formation at a depth of 895m, 92 bar and 33°C a high impurity  $CO_2$  stream with 15% non-condensables reduces the storage capacity by 39% compared with a pure stream of  $CO_2$ . It is, however, important to note that the quantity of storage available would be sufficient to provide storage for several centuries<sup>4</sup>, and therefore should not

<sup>&</sup>lt;sup>3</sup>Saline aquifers are likely to provide the largest storage capacity with an estimated global storage potential of 10,800  $\text{Gt}_{CO_2}$  [14] while depleted hydrocarbon reservoirs provide an estimated storage capacity of up to 1,000  $\text{Gt}_{CO_2}$  [15]

<sup>&</sup>lt;sup>4</sup>Current CO<sub>2</sub> annual emissions rate is at  $\sim 34 \text{ Gt}_{CO_2}$  [15]

be considered a limiting factor.

Table 1 shows the typical  $CO_2$  composition recommendations for  $CO_2$  transport and storage whilst only considering a source-to-sink approach where both recommendations - for transport and storage - are one. Certain impurities however may have a negative effect on transport and none on storage and vice versa. The presence of  $H_2O$  in the  $CO_2$  stream, for example, is very detrimental to the pipeline as it can cause corrosion. In addition,  $H_2O$  in the pipeline can result in hydrate formation which causes plugging [21]. Meanwhile,  $H_2O$  has no effect on storage efficiency [20] as it is already present in geological reservoirs.

#### 1.2. Oxy-combustion $CO_2$ capture

Oxy-combustion capture is currently one of the most promising methods of  $CO_2$  capture. It is one of the most developed so far for CCS, it allows for improved combustion efficiency [22] and can in principle be applied to any type of fuel used for thermal power generation [5]. The capture process, as illustrated in Figure 1, consists of separating oxygen from air, using an energy intensive Air Separation Unit (ASU), mixing the oxygen (and some excess nitrogen) with recycled flue gas and thereby providing a high-oxygen environment in which to burn the fuel of choice. This part of the system produces a saleable product, electricity, and a waste product, flue gas. The flue gas produced is rich in  $CO_2$  but still requires further purification and compression. Typical oxy-combustion flue gas compositions are presented in Table 1.

#### Table 1 about here.

The flue gas then goes through a  $CO_2$  Compression and Purification Unit  $(CO_2CPU)$  with a high energy demand and produces a dehydrated, high purity, high pressure  $CO_2$  product ready for transport and a waste product made up of impurities (H<sub>2</sub>O, O<sub>2</sub>, N<sub>2</sub>, Ar, SO<sub>2</sub>) and lost CO<sub>2</sub>.

#### [Figure 1 about here.]

Both the  $CO_2CPU$  and ASU impose approximately the same energy penalty on the power plant - each reduce the overall power plant efficiency by approximately 5% relative to the base plant efficiency [22]. Owing to the relatively impure raw exhaust gas which is produced from the oxy-combustion boiler, the  $CO_2CPU$  is crucial to providing a  $CO_2$  stream that is suitable for transport and subsequent sequestration. Thus far, work has been done on maximising the economic revenue of the ASU [23] and minimizing the energy penalty caused by the ASU [22, 24, 25, 26], while relatively little focus has been given to the cost of the  $CO_2CPU$  and how to drive this down. A few studies, however, have looked at modelling the purification and compression process of oxy-combustion derived flue gas for usage in EOR and for its sequestration [17, 27, 28]. This paper looks at the  $CO_2CPU$  part of the oxycombustion capture process considering a number of designs, each differing in the product gas purity, and looks at how this process can be manipulated to reduce the costs associated with oxy-combustion CCS.

#### 1.3. Objectives of this study

In the literature, the majority of studies consider a source-to-sink approach, particularly for  $CO_2$ -EOR [16, 19]. Although several studies have alluded to the benefits of having  $CO_2$  transport networks [6, 7, 11, 29], these have yet to demonstrate economic benefits that could stem from having such networks.

This study addresses the latter gap in literature by considering the combination of  $CO_2$  streams from various point sources into a transport network that leads to one major trunk line stream ready for injection. In order to do so, this is formulated as an optimization problem with a stringent lower bound on the final trunk line purity, minimizing cost and maximising purity. Such a scenario assumes that as long as each stream is dehydrated and compressed for transport, it does not need to reach injection-suitable purity before entering into the transport network, but only once the actual injection point is reached.

The remainder of this paper is laid out as follows. Section 2 presents and describes the four  $CO_2CPU$  models produced in Aspen HYSYS, their associated costs and system performances, and the investment model used to obtain a price for  $CO_2$ . Section 3 then presents the main contribution of this paper; demonstrating how a transport network system set up can reduce the cost of  $CO_2$  provided at the injection point whilst adhering to minimum purity requirement for storage.

# 2. Techno-Economic Analysis of Oxy-Combustion CO<sub>2</sub> Compression and Purification Units

The flue gas produced by oxy-combustion will vary in purity depending on a number of factors, including the type and quality of the fuel burnt, the amount of flue gas recycled for combustion and the water content (see Table 1). Therefore, the flue gas still requires dehydration, further purification and compression in order to be suitable for transport and storage. As discussed, the latter is performed by means of a  $CO_2CPU$ .

In view of the importance of the  $CO_2CPU$  within the oxyfuel combustion capture process, four variations of this process were modelled in Aspen HYSYS following previous work by Posch et al. [28]. These different models analyse the trade-offs that exist between purity of the  $CO_2CPU$  product and its cost, and are all based on typical phase separation techniques [17, 27, 28]widely used in carbon capture processes. The first  $CO_2CPU$  model is the most simple, consisting of a 6-stage compression and dehydration system only. As compression and dehydration is also required in post combustion capture, this model is applied to both scenarios: oxy-combustion flue gas and post combustion  $CO_2$  product 2. The three remaining models are built on the compression and dehydration model with increased complexity and different product purities. The first, in order of complexity, consists of a low purity double flash system without heat integration 5, followed by a double flash system with heat integration 4 and a high purity product and finally a CPU process with a 6-stage distillation column with incorporated heat integration 6. As these models increase in complexity, they increase in product purity and energy penalty while decreasing in separation efficiency.

These models use the Peng-Robinson equation of state [30] and mixing parameters of Kopke et al. 2008 [31]. This property method is suitable for the simple, non-associating fluids considered in the flue gas (CO<sub>2</sub>, H<sub>2</sub>O, N<sub>2</sub>, Ar, O<sub>2</sub>, SO<sub>2</sub>) and the range of temperatures (-60°C to 250°C) and pressures (1 bar to 120 bar) relevant to this study. Each plant was assumed to have 8460 working hours per year and a plant lifetime of 35 years. The flue gas inlet composition and flow rate are based on a pulverized coal firing power plant at nominal load [28]. These values are outlined in Table 2.

[Table 2 about here.]

# 2.1. Four models of the CO<sub>2</sub> Compression and Purification Unit Process 2.1.1. 6-Stage CO<sub>2</sub> Compression and Dehydration Train Model

This 6-stage  $CO_2$  compression and dehydration model is the least complex of the  $CO_2CPU$  models. The flue gas enters the system at 1 bar and 13°C and goes through a 3 stage pre-compression train with an inter-stage cooling and flash system. After each compression stage the flue gas exits at 10 bar, 20 bar and 30 bar in that order. All compressors are centrifugal with a polytropic efficiency of 85%. This applies to all compressors in the CO<sub>2</sub>CPU models discussed in this paper. Each inter-stage cooler, which uses cooling water at  $20^{\circ}$ C reduces the flue gas temperature to  $25^{\circ}$ C in order to avoid overheating of the system as the gas is compressed. The fluid then goes through a flash after each compression-cooling stage. Once the flue gas has reached 30 bar, it is sent through a dehydrator. After each 3 stage precompression the flue gas is then taken through a dehydration system, which is described in more detail in section 2.2.3. Finally, the dry fluid exiting the dehydrator is compressed to 60 bar, cooled to 25°C compressed again to 80 bar, cooled to 15°C using refrigerant propane and compressed a final time to 120 bar - a fluid pressure suitable for pipeline transport [2]. In this study. we assume that at 120 bar and above, the  $CO_2$  streams considered are all supercritical (well above the critical point).

#### [Figure 2 about here.]

This process gives a  $CO_2$  product at 82.9 wt% purity. It is important to note that the composition of the  $CO_2$  product stream obtained will vary with the composition of the inlet stream.

In the post combustion  $CO_2$  capture process a compression and dehydration unit is also required to eliminate the remaining water in the stream and obtain a pressure adequate for transport. This process model is represented in figure 2 and its respective inlet and outlet conditions are highlighted. The characteristics of the  $CO_2$  stream entering the system are shown in Table 3. The wet  $CO_2$  stream enters the  $CO_2CPU$  at 98.5wt%  $CO_2$  and exits at 99.97wt.% purity.

[Table 3 about here.]

The post combustion stream entering the  $CO_2CPU$  is at a pressure of 2 bar - as opposed to 1 bar in the oxy-combustion system - which is the typical pressure for the stripper/solvent regeneration column in amine-based post combustion [22, 32]. In the model, the post-combustion stream first enters into a flash prior to the first compressor and cooler. This is due to the fact that a fraction of the stream entering the system is in aqueous phase. The rest of the pre compression train then follows the same pattern - the first compressor takes the stream up to 10 bar, the next to 20 bar and the third to 30 bar - with the exception of the last cooler and flash combination. The installed cost per tonne of  $CO_2$  captured yearly for different process units (compressors, pump and coolers) and the electricity cost per tonne of  $CO_2$  captured are illustrated in Figure 3. Cooler 3 referenced in Figure 3 only applies to 3-stage pre-compression train of the oxy-combustion flue gas stream and is not present when applied to the post-combustion flue gas stream (see Figure 2).

#### [Figure 3 about here.]

From the results obtained, we distinguished between the proportion of the difference in system cost due to the post combustion stream coming in at 2 bar. An additional  $\pounds 2.5/t_{CO_2}$  processed would be added to the installed cost of compressor 1 if the inlet stream pressure were at 1 bar. An additional  $\pounds 1.8/t_{CO_2}$  captured yearly would be added to the annual electricity costs if the inlet stream of the post combustion CPU were at 1 bar as opposed to 2 bar. This implies that the remainder of the difference in electricity costs between the post combustion and oxy-combustion  $CO_2CPUs$  (£1.8/t<sub>CO2</sub> captured yearly) is due to the higher amount of impurities in the oxy-combustion process streams. A recent study by the IEA GHG R&D Programme explains that the larger presence of non-condensable gases with critical temperatures and pressures below that of  $CO_2$  shift the two phase region upwards [20]. These non-condensable gases then take up a larger volume of the fluid and more work is required for compression. The use of a pressurized combustion process for oxy-combustion could therefore substantially reduce both the capital and operating costs associated with the first compressor of the  $CO_2CPU$ . However, compressing high purity oxygen for combustion in turn has very high cost implications due to the special materials that would be required for the compressor and piping to avoid oxidation [33]. A number of operational and safety issues would also need to be considered when designing a

combustion chamber for oxy-combustion as described in the IEAGHG report on Oxy Combustion Processes for  $CO_2$  Capture from Power Plant [34]. The remaining difference in installed cost of compressors for each system results from the difference in density between the two streams.

Furthermore, coolers 4 and 5 have higher installed cost per tonne of  $CO_2$  captured in the post combustion system. This is due to the molar enthalpy of the stream going through coolers 4 and 5 being much lower in the post combustion system than it is in the oxy-combustion system thereby requiring more heat exchange surface. Finally, coolers 1 and 2 have very small installed costs per tonne of  $CO_2$  captured (between  $\pounds 0.05/t_{CO_2}$  and  $\pounds 0.06/t_{CO_2}$ , while cooler 3 is only present in the oxy-combustion version of this  $CO_2CPU$ .

#### 2.1.2. High Purity Double Flash CO<sub>2</sub>CPU Model with Heat Integration

A second model of the  $CO_2CPU$  is developed with increased complexity. This model brings in the same flue gas as described in Table 2 and takes it through a 3 stage pre-compression train with an inter-stage cooling and flash system. As in the previous model, the flue gas is compressed to a pressure of 30 bar and the flue gas is sent through a dehydrator as described in section 2.1.1. The dehydrated and compressed flue gas is cooled down to -27°C via multi stream heat exchanger 1 and then goes into flash 1 (see figure 4). Out the bottom of flash 1, a  $CO_2$  rich stream is expanded in an adiabatic throttle and is used as a cooling agent in multi stream heat exchanger 1. The top product from flash 1 is the  $CO_2$  poor stream with a  $CO_2$  mass fraction of 0.67. This stream is recycled and cooled in multi stream heat exchanger 2 to  $-54^{\circ}$ C before going through flash 2. The CO<sub>2</sub> product out the bottom of flash 2 remains at approximately -54°C and is used as a cooling agent in multi stream heat exchanger 2. This stream is then expanded through an adiabatic throttle to 8 bar and recycled through multi stream heat exchanger 2 and again through multi stream heat exchanger 1. The cooling potential of the top gaseous products of flash 1 and flash 2 are also recycled through multi stream heat exchangers 1 and 2 as illustrated in figure 4.

Finally the bottom products from flash 1 and flash 2 are mixed together, forming a high purity  $CO_2$  stream at 8 bar and  $-10^{\circ}C$ . The product stream then goes through a two stage compression (60 bar then 80 bar) with interstage cooling, bringing the stream down to 25°C after each compression, then pumped up to 120 bar. The final product stream achieves a  $CO_2$  purity of 97.5 wt.%.

The waste stream from the  $CO_2CPU$  system is the top product of flash

2 with remaining cooling potential at 30 bar and -35  $^{\circ}\mathrm{C}$ . At appropriate temperature and pressure, this waste stream could be used as a flue gas recycle into the combustion chamber.

#### [Figure 4 about here.]

A range of temperatures and pressures were tested for the system bearing in mind that the triple point of  $CO_2$  is at 5.19 bar and -56.6°C [35]. The temperature and pressures outlined above were found most suitable for obtaining a high purity  $CO_2$  stream whilst minimizing compression and cooling duty as described in the work by Posch et al. [28].

#### 2.1.3. Low Purity Double Flash $CO_2CPU$ Model without Heat Integration

A similar double flash  $CO_2CPU$  system was also modeled in Aspen HYSYS using much higher pressure (68 bar after the upstream compression train) and higher temperature (-18°C stream coming into Flash 1). The operating pressure of 68 bar was chosen because at that pressure and very low temperatures (between -55°C and -44°C) a mixture of  $CO_2$ , N<sub>2</sub> and O<sub>2</sub> was separated into a liquid and a vapour phase with most of the  $CO_2$  in the liquid phase and most of the N<sub>2</sub> and O<sub>2</sub> in the gas phase [36]. Since Ar behaves very similarly to N<sub>2</sub> and O<sub>2</sub> at such pressure and temperature [20] we use the latter operating pressure and temperature in our Aspen HYSYS model.

Once the flue gas is compressed to 68 bar in the pre-compression train, dehydrated then cooled to  $-18^{\circ}$ C it then goes through a first flash with a CO<sub>2</sub>-rich bottom stream at 92.7wt.% purity. The top product, still containing 15% of the CO<sub>2</sub> in the system, is cooled to  $-55^{\circ}$ C and taken through a second flash that produces a CO<sub>2</sub> rich bottom product at 91.3 wt.% CO<sub>2</sub> and a waste stream top product with some remaining CO<sub>2</sub>. Each of the bottom product CO<sub>2</sub> streams are taken through adiabatic throttles and expand to 10 bar at which point they mix and form a stream of 92.6wt.% CO<sub>2</sub> and are compressed up to 120 bar in the final compression stage.

#### [Figure 5 about here.]

#### 2.1.4. CO<sub>2</sub> Compression and Purification Unit with a 6-Stage Distillation Column

The third  $CO_2CPU$  model represents the most complex system. The 3stage pre compression train 6 is identical to that in the 6-stage compression and dehydration unit (see figure 2) with the exception of the third compressor that brings the flue gas to a pressure of 28 bar. After going through the pre-compression stage, the flue gas is sent to a dehydrator as specified in section 2.1.1. Once the stream is dry it goes through the multi-stream heat exchanger where it is cooled to  $-35^{\circ}$ C. This stream then goes through the 6 stage distillation column, comprising of a partial condenser with flash 1 at the top and a partial reboiler with flash 2 at the bottom. Flash 1 splits the top product into a vapour fraction of 0.45, whereas flash 2 splits the bottom product into a vapour fraction of 0.75. Six stages for the distillation column was found to be optimum in giving a good trade-off between the high purity of the CO<sub>2</sub> stream coming out as the liquid bottom product and the higher flow rate of CO<sub>2</sub> coming off the top of the distillation column [28].

The top vapour product passes through the multi stream heat exchanger and is cooled to  $-55^{\circ}$ C and then the vapour-liquid mixture is separated in flash 3. The CO<sub>2</sub>-rich bottom product from flash 3 passes through an adiabatic throttle where it is expanded and cooled to  $-55.5^{\circ}$ C and its cooling capacity used through the multi stream heat exchanger, then compressed back up to 28 bar and cooled down to  $-35^{\circ}$ C before being recycled back into the inlet stream of the distillation column. The top product of Flash 3 (the waste stream) is expanded and cooled through another adiabatic throttle to  $-66^{\circ}$ C and its cooling capacity used through the multi stream heat exchanger. This stream contains only 25mol% CO<sub>2</sub> and the remainder is a combination of O<sub>2</sub>, N<sub>2</sub> and Ar. Due to the very low freezing points of the latter components, -219°C -210°C and -189°C respectively, the freezing point of the mixture is well below  $-66^{\circ}$ C [37].

The CO<sub>2</sub>-rich distillation column's bottom liquid stream then passes through an adiabatic throttle where it is expanded and cooled to  $-54^{\circ}$ C and is used for its cooling potential through the multi-stream heat exchanger, before being sent through to the final compression stages and pumped up to 120 bar, 36°C and achieving a final CO<sub>2</sub> product purity of 99.98 wt.%.

#### [Figure 6 about here.]

The choice of temperature and pressure going into the distillation column were based on minimising compression duty whilst maximising cooling capacity circulated in the system. Posch et al. [28] found that entering the distillation column at 28 bar was best in satisfying cooling agent demand and entering at a temperature of  $-35^{\circ}$ C was optimal for minimising compression duty in the system.

#### 2.2. CO<sub>2</sub>CPU System Performance

#### 2.2.1. Product Purity, System Efficiencies, CAPEX and OPEX

Having described the difference in complexity, units and product of each of these four models, this section presents the economic evaluation and system performance for each of the models. The economic evaluations were performed using the installed Aspen HYSYS economics tool and a UK metric cost option was incorporated. The default utility assumptions used in Aspen HYSYS are presented in the Appendix 6.1. Given the extensive AS-PEN Tech database on unit operations and cost, we assume that these are at the nth of a kind stage (NOAK). The economic evaluations and financial analysis that follows are given at a constant 2015 cost level [38, 39].

The separation efficiency of each system was calculated using Equation 1. The separation efficiency is the amount of  $CO_2$  that is captured in the product stream over the amount that comes into the CPU system.

$$\eta_{sep} = \frac{\dot{m}_{CO2,product}}{\dot{m}_{CO2,inlet}} \tag{1}$$

where:

$\eta_{sep}$	= Separation efficiency of $CO_2$ from the initial flue gas into
	the system
$\dot{m}_{CO2,product}$	= Mass flow rate of $CO_2$ out of the system (after product
	compression for storage)
$\dot{m}_{CO2,inlet}$	= Mass flow rate of $CO_2$ into the system (before
	pre-compression stage)

The results in Table 4 show that as the models increase in complexity and product purity (descending order of complexity: Distillation > Double Flash with Heat Integration > Double Flash without Heat Integration > Compression and Dehydration), the capital costs (CAPEX) and operational costs (OPEX) incurred increase as well. This is due to an increase in energy requirement per tonne of  $CO_2$  captured and an increase in power plant net efficiency loss with increased complexity and product purity.

Notably, the 6-stage Distillation CPU model with a capital cost of £51

million presents a much higher cost burden than the other  $CO_2CPU$  models. This is primarily due to the presence of a distillation column. The double flash system without heat integration was found to have the second highest capital cost at £42 million which is mainly due to the larger compressors required upstream (compression to 68 bar) and the large heat exchange areas required for the interstage cooling upstream. In addition, the operating costs incurred for the distillation CPU and the double flash with heat integration CPU systems are substantially higher than for the  $CO_2$  compression and dehydration unit, £6 million and £4 million higher, in spite of starting and ending at the same pressures.

In addition, the power plant net efficiency loss (in percentage points) caused by each type of  $CO_2CPU$  was calculated assuming that the power plant output without the capture plant was 347 MWe and that the average efficiency of a pulverised fuel coal fired power plant is of 40% (LHV basis) [40].

The main process results, capital and operational costs for all process models, including the CPU for a post combustion derived flue gas are presented in Table 4. In order to verify our results we compared them with other oxy-fuel purification process models with similar product streams presented in literature. White et al. [27], for example, finds that for an oxy-fuel CO<sub>2</sub> purification system taking a CO<sub>2</sub> stream from 1 to 110 bar and achieving a purity of 99.97 mol% CO<sub>2</sub>, 177 kWh/tonne of CO<sub>2</sub> captured are consumed. Our results for the highest purity system deviates by  $5kWh/t_CO_2$  (see Table 5).

As the separation efficiency increases, the amount of work required per tonne of  $CO_2$  captured decreases with it. However, as the product purity decreases, the power required first decreases before increasing again with the double flash without heat integration system, then the power required decreases again while increasing the amount of  $CO_2$  captured. This is consistent with the results presented by White et al. [27]. Hence, while the systems decrease in product purity and complexity, they increase in separation efficiency. The compression and dehydration system captures 100% of the  $CO_2$ in the system, whereas the Double Flash without heat integration and with heat integration capture 97% and 92% of the  $CO_2$  entering the system as a raw flue gas. The 6-stage Distillation CPU process captures only 90% of the  $CO_2$  entering the system. Hence, with decreased system complexity, less  $CO_2$ would be vented to the atmosphere and indeed more of the  $CO_2$  coming into the system would be captured. In a context in which unabated  $CO_2$  would be penalised in the UK and other countries, a lower separation efficiency may also imply a higher system cost.

These results indicate that trade offs will need to be made between purity and capture efficiency, large capital cost investment risks resulting in high purity products and lower capital investment risk resulting in lower potentially less suitable product purity. In turn, the latter presents the trade off between high power plant net efficiency loss, higher product purity, higher electricity costs and lower power plant net efficiency loss, lower product purity, lower electricity costs.

The system costs for the compression and dehydration unit applied to a typical amine-based post combustion stream are significantly lower than that of the same process model used for an oxy-combustion derived stream, with a capital cost of £23 million and £33 million respectively and an £11 million difference in yearly operational costs. As discussed in section 2.1.1 and shown in Figure 3, this is a result of the difference in inlet pressures as well as the larger amount of non-condensable gases present in the oxy-combustion derived stream causing additional compression work.

[Table 4 about here.]

Table 5 about here.

The next section, section 2.2.2, puts these costs into perspective and interprets them as a marketable  $CO_2$  price.

#### 2.2.2. An Investment Decision

In order to put these results into perspective a  $CO_2$  price was established for each of the resulting process model product streams discussed above, assuming there is a market for  $CO_2$  in the EOR sector. The approach taken was to assume that the  $CO_2CPU$  is an entity independent of the power plant and the ASU, as represented in Figure 1. This implies that the cost of the ASU impacts the cost of electricity output from the power plant whilst the cost of the  $CO_2CPU$  does not. We assume that as an independent entity, the  $CO_2CPU$  has two inputs - a cost-free raw material (flue gas from the power plant) and a set of expenses (energy cost) - and two principal outputs - a product/revenue stream ( $CO_2$  at high pressure and purity) and a waste stream consisting of a gas mixture ( $CO_2$ ,  $N_2$ ,  $O_2$  and Ar) and water. Hence, in order to sustain this business approach, we established a price for the  $CO_2$  product from each of the  $CO_2CPU$  process options assuming that the investments in the  $CO_2CPU$  would only be made if the minimum rate of return were of 20% post tax. A minimum required rate of return of 15%-20% (post-tax) is deemed appropriate for an early-stage project. As the maturity of the project increases the latter can be reduced to 5%-10% [29]. This analysis assumes that a lower purity  $CO_2$  stream would have the same market share as a higher purity  $CO_2$  stream.

Using the results produced by the Investment Analysis function in Aspen HYSYS, we lay out the balance sheet for each unit. In our analysis we assumed a debt to equity ratio of 60:40, a working capital of 5% and an interest rate of 10%. As a result we obtained a  $CO_2$  product stream price for which the internal rate of return (on the  $CO_2CPU$  only) matches the minimum rate of return (also called the hurdle rate) of 20%, indicating that the investment would go through. The results from this analysis are presented in Figure 7.

As observed in Figure 7 the relationship between  $CO_2$  price and stream purity is non-linear and non-monotonic. The non-monotonicity is due to the fact that the double flash system without heat integration is highly inefficient and requires a much higher pressure state in order to obtain a separation that gives a 92.6wt.% purity. The  $CO_2$  price resulting from this  $CO_2CPU$  would also decrease if heat integration were applied. The non-linearity observed is entropically driven: as we obtain a higher purity stream the incremental increase in purity becomes harder and more costly to achieve [27, 28]. For the remainder of this study, the double flash without heat integration CPU will be excluded as it has shown to be more costly than alternative systems with higher product purities. The same analysis is also applied to the CPU used for post combustion and, assuming that the CPU is an independent entity in which to invest - separate in this case from the post combustion amine-based process - a  $CO_2$  price of £8.9 per tonne is obtained for a product stream of 99.97 wt.% purity. It is important to note that the CPU needed for postcombustion capture is only a part of a cost intensive capture process (1/3 of)the energy penalty resulting from post-combustion capture [22]) and that the price obtained does not reflect the other cost intensive capture processes.

The results show that for the highest purity achievable from the  $CO_2CPU$ , a saleable  $CO_2$  stream at a price of £19.8 per tonne of  $CO_2$  would make the investment worth-while whereas a  $CO_2$  stream that is saleable at a purity of 82.9 wt.% would only need to be sold for £11.4 per tonne of  $CO_2$  in order to make the investment in the  $CO_2CPU$  lucrative. This difference represents a 42% decrease in product price. However, none of these results are meaningful unless a market for  $CO_2$  exists and here we assume that the market buyers would be oil producers using  $CO_2$  for EOR. As EOR can provide important stimulation for CCS deployment by reducing the overall costs of  $CO_2$  capture and transport, thereby reducing the investment risk [41], it is critical to attract this EOR market with a low  $CO_2$  price [29]. Although  $CO_2$ -EOR offshore has yet to be done in the North Sea, studies have shown the feasibility and benefits that could be brought on by its deployment [42, 43, 44, 45].

However, if CO<sub>2</sub> for EOR maintains strict requirements in terms of the stream purity and the oxygen content (limited to 100 ppm), this eliminates the possibility of all product streams with the exception of the 99.98wt.% purity one at  $\pounds 19.8/t_{CO_2}$  to be used for EOR. Nevertheless, as discussed in Section1.1, there is leverage to relax CO<sub>2</sub> requirements for use in EOR and thereby consider a lower purity and cheaper CO<sub>2</sub> stream. In addition, if a combination of the cheaper lower purity sources of CO<sub>2</sub>, at purities 82.9wt.% and 97.5 wt.% for  $\pounds 11.4/t_{CO_2}$  and  $\pounds 16.5/t_{CO_2}$ , were to be mixed with inherently higher purity sources of CO<sub>2</sub> (e.g. from post combustion capture or natural gas processing) in a transport network system, the aggregation of streams could produce a stream suitable for injection and EOR at a much lower overall cost.

The same method is applied assuming minimum rates of return of 15%, 10% and 5% and the resultant CO<sub>2</sub> prices are represented by the dashed and dotted lines in Figure 7. As expected, the CO<sub>2</sub> price decreases as the minimum rate of return required decreases. However, the reduction in price is greatest for the streams with highest capital cost: the 6-stage Distillation CPU and the Low Purity Flash CPU. This implies that as a CCS industry becomes more established, and less exposed to risk, minimum rate of return could be decreased, allowing one to further reduce costs. We compare these prices to studies of the cost of CCS summarized in the review paper by Rubin et al. 2015 [46]. The latter study quotes oxy-combustion capture cost to be between  $24\pounds/t_{CO_2}$  and  $45\pounds/t_{CO_2}$  at a 2015 cost level. Considering that the CO<sub>2</sub>CPU accounts for approximately half of the oxy-combustion capture costs, we conclude that our price values fall in line with this range.

### [Figure 7 about here.]

#### 2.2.3. Triethylene Glycol Dehydration System

The models and cost analysis described above considers a dehydration system that acts as a 'black box' separation system comprising only of operational costs involved in stripping the respective streams of water and is not associated with an equipment cost. For completeness of this study, a triethylene (TEG) glycol  $CO_2$  absorption system is modelled in Aspen HYSYS using the HYSYS glycol property package and its cost assessed using the Aspen HYSYS economic evaluation tool. Absorption was chosen over adsorption as adsorption dehydration plants typically have higher installation costs and glycols (used in absorption) are cheaper than adsorbents and easier to replace than an adsorber bed [47, 48, 49]. Furthermore, triethylene glycols are chosen amongst other glycols as these are best for gas dehydration as they can be regenerated at high concentration without degradation at high temperatures due to their high degradation temperature ( $260^{\circ}$ C) [48, 50, 51]. The dehydration system (illustrated in Figure 8) consists of an absorber that pumps in TEG to the pressure of the inlet flue gas stream (either 30bar or 28 bar see Section 2.1) at the top and water-rich flue gas entering at the bottom of the column. The dry flue gas then exits the absorber as the top product (with less than 10ppm  $H_2O$ ) and water-rich TEG as bottom product. The TEG stream expands through a throttle value to atmospheric pressure and through a flash vessel then splits off a small amount of waste flue gas. The TEG stream is then heated up to  $150^{\circ}$ Cand enters the stripper for regeneration of the TEG stream. The dry TEG stream leaves the bottom of stripper and is cooled using propane as a refrigerant before being recycled back into the dehydration system. The costs associated with this dehydration system are shown in table 6. It is assumed that fresh TEG is purchased once yearly given a usage rate of 2000kg/hour and at  $\pounds 1.57/\text{kg}$  [52]. Due to the pressure at which the absorption occurs, we do not apply this dehydration system to the Low Purity Double Flash model (described in section 2.1.3) and ignore the latter  $CO_2CPU$  model for the rest of the study as it is deemed uneconomical.

[Figure 8 about here.]

[Table 6 about here.]

#### 3. A CO<sub>2</sub> Transport Network System

#### 3.1. Transport Network Scenario

Having a number of purification options at different costs is worth exploiting when applied to a number of sources with various capture technology options to drive down overall costs of capture in a given region. In order to demonstrate this point we develop a  $CO_2$  transport network based on capture rates from real UK power and industrial plants inputting  $CO_2$  from different sources and with varying product purities. We use a UK-based case study by Prada et al. [53] to get representative flow rates from 10 combined cycle gas turbine (CCGT) plants, 10 coal fired power plants and 1 steel plant. The flow rates from these  $CO_2$  outputs and their hypothetical capture plants are shown in Table 7. Each plant is then coupled with a type of capture plant which will in turn give the purity of the  $CO_2$  stream extracted from a given power or industrial plant. Here, we consider the prices obtained for each product stream from the different  $CO_2CPUs$  described in Section 2. CCGT plants are constrained to exclusively adopt post combustion capture. This is due to the current absence of gas turbines designed for oxy-combustion. The flue gas composition resulting from post combustion capture, prior to entering the CPU, is given in Table 3. The coal power plants and the steel plant are assumed to adopt oxy-combustion  $CO_2$  capture. In the case of  $CO_2$  capture from a steel production plant, oxy-combustion - as opposed to post-combustion - provides the flexibility needed to make up for the varying compositions of flue gas from the blast furnace as well as the high temperature flue gas [54]. Each of the  $CO_2$  sources considered are assumed to join at a central hub from which a trunk line of  $CO_2$  brings the combination of streams to the Bacton terminal and sent through for storage in the Southern North Sea. This scenario is illustrated in figure 9.

#### [Figure 9 about here.]

This scenario was formulated as a bi-objective optimization problem examining the trade-offs between optimizing for least cost and greatest purity. The problem is formulated as a mixed-integer linear programming model (MILP). The resulting MILP model was solved using the GAMS CPLEX solver as this is well suited for solving problems of this type [55]. This work shows that, for a minimum purity required at the injection point, mixing high purity sources of  $CO_2$  with lower purity sources of  $CO_2$  within the transportation network can significantly reduce the cost of the final  $CO_2$  product stream for injection and thereby improve the economic viability of the system.

#### [Table 7 about here.]

3.2. Formulation of the Optimization Problem

The optimisation problem is expressed as follows:

$$[h!]\min[y(i)], subject to x(i) \ge 0.96,$$
(2)

$$[h!]\min[z(i)], subject to x(i) \ge 0.96, \tag{3}$$

where y and z, the objective functions we seek to minimize, are the capital cost incurred per tonne of CO<sub>2</sub> captured, respectively and the operational cost incurred per tonne of CO<sub>2</sub> captured. x is the final product purity at the trunk line ready for injection.

The optimization variables are the choices of  $CO_2CPU$  systems *i* and include a post-combustion CPU option (PostC) in subset *v*, and three oxycombustion CPU options in subset *u*: a distillation unit (Dist), a high purity double flash unit (HPDF) and a compression and dehydration unit (CD). In this problem we do not consider variations of post combustion capture options and instead assumes that the  $CO2_2CPU$  processes a typical amine-based post combustion captured  $CO_2$  stream at 90% capture efficiency of purity of 99.97 wt.% [53]. After going through the compression and dehydration unit modeled in section 2.1.1 the composition of the final post combustion  $CO_2$ stream is given in Table 4.

All of the optimisation parameters and variables are described in Appendix 6.2. For each capture plant option a number of optimization parameters are given based on the assumptions made for post combustion capture and the costs derived in section 2.2 for the compression and purification units.

For the purpose of this study, we only take into account the capital and operational costs that stem from the  $CO_2$  Compression and Purification Units needed for oxy-combustion and the compression and dehydration unit needed in the final stage of post combustion capture. Hence, when talking about an oxy-combustion option or a post combustion option this refers solely to the choice of  $CO_2CPU$ . With each category of capture and subset of CO<sub>2</sub>CPU option several optimization parameters are given. These include: CO<sub>2</sub> product purity resulting from each oxy-combustion option A1(u)and resulting from the post combustion option A2(v), capital cost in  $\pounds/t_{CO_2}$ captured per year for oxy-combustion options  $CAP\_oxy(u)$  and for post combustion  $CAP\_post(v)$ , operational costs  $\pounds/t_{CO_2}$  captured per year for oxy-combustion options  $OP\_oxy(u)$  and for post combustion  $OP\_post(v)$ .

Each plant is separated into the three categories as described in section 3, with j referring to the gas plants, k the coal plants and l the industrial plant (one steel plant only in this example). The Flow rate for each of the plants are given by the following optimization parameters:  $Flow\_gas(j)$ ,  $Flow\_coal(k)$ ,  $Flow\_ind(l)$ .

We introduce two binary variables in order to allow for the plants to choose among the different discrete  $CO_2CPU$  options. These binary variables are decision variables that take integer values only, hence requiring the use of a mixed-integer linear programming (MILP) solver for this problem. We introduce equations 4 and 5 that the binary variables are constrained by in order for each plant to select only one  $CO_2CPU$  option:

$$[h!]\sum_{u}binary1(u,k) = 1 \quad \forall k \tag{4}$$

$$[h!]\sum_{u} binary2(u,l) = 1 \quad \forall l \tag{5}$$

These binary variables are used to calculate the product purity, the capital and operational costs for the sum of all coal streams and for the product resulting from the capture of  $CO_2$  from the industrial plant. The equations set up to calculate the final product purity, capital cost and operational cost per tonne of  $CO_2$  in the trunk line ready for injection are given in Appendix (6.2). The constraint set on this problem is for the final trunk line purity, x, of at least 96 wt.% for injection with ppm levels of water. As a result of this study we obtain the quantitative trade off between the two conflicting objectives: minimum cost and maximum purity.

Figure 10 represents the bi-objective optimisation Pareto front for the competing objective functions: minimum cost and maximum purity. It is clear that these are competing given that by increasing product purity, the cost will increase as well. The purity of the  $CO_2$  stream in the trunk line, x, has a lower bound set at 0.96 mass fraction. The first solution, to the far left

represents a scenario in which the West Burton coal plant (see Table 7) uses the CD  $CO_2CPU$  and the rest of the coal plants and the steel plant use the HPDF  $CO_2CPUs$ . As explained in section 3, the gas plants are assumed to adopt the post combustion capture method and therefore contribute a fixed cost and purity in all scenarios. This first result gives a minimum capital cost of  $\pounds 13.83/t_{CO_2}$  and a minimum operational cost of  $\pounds 12.54/t_{CO_2}$  for minimum acceptable mass fraction of  $CO_2$  in the trunk line of 0.96. As we increase the minimum purity demanded at the injection point the capital cost increase follows a non-linear trend. At a minimum mass fraction of  $CO_2$  of 0.98 (see Figure 10) the slope increases significantly, representing a scenario in which all coal and steel plants use an HPDF CPU and Tilbury, a coal plant, has just switched from an HPDF CPU to a Dist CPU. This scenario gives a capital cost of  $\pounds 14.45/t_{CO_2}$  and operational cost of  $\pounds 12.99/t_{CO_2}$ . Once, the minimum mass fraction of  $CO_2$  in the trunk line reaches 0.992, the slope of the CAPEX curve sees a sharp increase before reaching a plateau at 0.995  $CO_2$  mass fraction. The plateau reached, represents a scenario in which all coal and industrial plants have switch to a Dist CPU system reaching a capital cost of  $\pounds 21.03/t_{\rm CO_2}$  and operational cost of  $\pounds 14.17/t_{\rm CO_2}$ .

#### [Figure 10 about here.]

Figure 10 shows that above a certain purity requirement of approximately 98 wt.% CO<sub>2</sub>, the capital investment cost increases drastically, increasing the investment risk and CO<sub>2</sub> price. This steep increase in cost when aiming for a purity above 98 wt.% CO<sub>2</sub> is also reflected in the CO<sub>2</sub> price curve shown in 7 as the above design point of the CO<sub>2</sub>CPU is set at a much higher capital cost to achieve 99.98 wt.% CO<sub>2</sub>. Using the investment analysis presented in section 2.2.2 and the prices that reflect the cost of each CO<sub>2</sub>CPU with a different product stream - applied both to oxy-combustion and post combustion - a final trunk line product stream price is obtained for the two ends of the Pareto front curve. In the 20% minimum rate of return on investment scenario, when optimising for minimum cost, we obtain a final trunk line CO<sub>2</sub> stream at 96.8wt.% purity for a price of £14.68/t<sub>CO2</sub>. Whereas, when optimising for maximum purity we obtain a final trunk line stream of 99.98wt.% purity at a price of £17.77/t<sub>CO2</sub>. This difference represents a 17.4% saving in CO<sub>2</sub> product price.

As the minimum rate of return required is reduced to 15%, 10% and

5% the savings resulting from having the lowest cost stream as opposed to the highest purity stream are reduced. These results are shown in Figure 11. This implies that as the maturity of the technology increases and the required minimum rate of return therefore decreases, the trade-offs between having a high purity stream at higher cost versus a lower purity stream at lower cost will be less significant. These results are valuable both on the capture side as well as the  $CO_2$  market side. For iron, steel and cement production, it is crucial to be able to provide a low cost  $CO_2$  capture option in order to provide such plants with a low-carbon option that is more worthwhile than relocating to a region with less stringent emissions reduction policies. In being able to mix low purity  $CO_2$  streams with higher purity ones, producing a final stream suitable for injection, this low cost scenario is rendered possible for such industries. As for the  $CO_2$  market, considering a context in which CO<sub>2</sub>-EOR is desired, the process would only be economical at a reasonable and low  $CO_2$  price, if any at all. This study, however, does not include transportation costs and therefore does not take into account the fact that having a cheap, low purity  $CO_2$  stream will increase the required pipeline diameter and thereby increase the transportation costs incurred [11]. Nonetheless, this has been shown to have little or no effect on pipeline costs over short distances (30km), and this is one of the key advantages of using a multi-hub transport network as in this study [4].

[Figure 11 about here.]

#### 4. Conclusion

As the need for decarbonising the power and industrial sectors becomes increasingly apparent, CCS stands out as a crucial means for reducing greenhouse gas emissions. With this comes the demand for economic incentives to deploy CCS such as using CO<sub>2</sub> for EOR. In order to take advantage of these potential markets and maximise the financial return it is crucial to minimise the costs involved in capturing the CO<sub>2</sub>.

Through a detailed study of four  $\text{CO}_2$  compression and purification units applicable to oxy-combustion capture we have shown that higher  $\text{CO}_2$  purity is associated with higher costs of capture and lower capture efficiencies. This was translated into a  $\text{CO}_2$  price, assuming that the  $\text{CO}_2\text{CPU}$  is a business unit independent of the rest of the oxy-combustion plant. The price observed points out that the capture plant could, on one extreme, sell a lower purity dehydrated  $\text{CO}_2$  stream (82.9 wt.%  $\text{CO}_2$ ) to the market for 42% less than the capture plant producing the highest purity  $\text{CO}_2$  (99.98 wt.%  $\text{CO}_2$ ). However, the cheapest  $\text{CO}_2$  product stream is not suitable for injection into a storage site or to use for EOR, but is suitable for transport.

An important conclusion from this work is that applying different capture options - providing  $CO_2$  at different prices and purities - to a network of capture plants and transport infrastructure has the potential to substantially reduce the cost of the  $CO_2$  capture and transport system, whilst achieving a final  $CO_2$  product that is suitable for injection. This is achieved by combining several streams of  $CO_2$  at different purities into one final trunk line that then delivers the product to the storage sink. The costs considered only pertain to the  $CO_2$  compression and purification units.

It is important to point out that, as opposed to the US, where  $CO_2$ -EOR common practice takes  $CO_2$  from one natural source with high purity  $CO_2$  and transports it to one sink, new  $CO_2$ -EOR projects must look at minimising cost to make up for the high risk investment.

This work showed that a 17.4% reduction in  $CO_2$  price can be obtained by combining multiple  $CO_2$  sources into one transport network, taking  $CO_2$ from post combustion plants and oxy-combustion capture process CPU options and inputting these into one final trunk line. It is interesting to note that this price difference is obtained when assuming a 20% minimum rate of return on investment, which is what is typically demanded of a high risk investment. However, as more such plants and networks are deployed, the investment risk will decrease, thereby the minimum rate of return required will decrease and as a result the gap in price of the  $CO_2$  resulting from the maximum purity system and the minimum cost system narrows.

### 5. Acknowledgements

The authors would like to thank the UK Natural Environment Research Council (NERC) for funding this research.

#### 6. Appendix

#### 6.1. Process Utility Assumptions

[Table 8 about here.]

#### 6.2. Transport Network Optimisation Problem

This appendix presents the equations and constraints that give the objective functions x (product purity), y (CAPEX in  $\pounds/t_{CO_2}$ ) and z (OPEX in  $\pounds/t_{CO_2}$ ). The following set of equations give the purity of the sum of CO<sub>2</sub> streams from each type of plant, the capital cost incurred per tonne of CO<sub>2</sub> captured for the sum of each type of plant and the operational cost incurred per tonne of CO<sub>2</sub> captured for the sum of each type of plant in the following order: gas CCGT plants, coal plants, industrial plants (one steel plant). The variables for each equation are defined in Table 9.

$$\begin{split} \sum_{v} [Flow\_gas(j) * A2(v)] &= CO2\_1(j) \quad \forall j, \\ \sum_{u} [Flow\_coal(k) * A1(k) * binary1(u,k)] &= CO2\_2(k) \quad \forall k, \\ \sum_{u} [Flow\_ind(l) * A1(u) * binary1(u,l)] &= CO2\_2(l) \quad \forall l, \\ \sum_{v} [Flow\_aga(j) * CAP\_post(v)] &= CO2CAPEX1(j) \quad \forall j, \\ \sum_{v} [Flow\_coal(k) * CAP\_oxy(u) * binary1(u,k)] &= CO2CAPEX2(k) \quad \forall k, \\ \sum_{u} [Flow\_ind(l) * CAP\_oxy(u) * binary1(u,l)] &= CO2CAPEX3(l) \quad \forall l, \\ \sum_{v} [Flow\_aga(j) * OP\_post(v)] &= CO2OPEX1(j) \quad \forall j, \\ \sum_{v} [Flow\_coal(k) * OP\_oxy(u) * binary1(u,k)] &= CO2OPEX1(j) \quad \forall j, \\ \sum_{v} [Flow\_aga(j) * OP\_oxy(u) * binary1(u,l)] &= CO2OPEX2(k) \quad \forall k, \\ \sum_{v} [Flow\_ind(l) * OP\_oxy(u) * binary1(u,l)] &= CO2OPEX3(l) \quad \forall l, \\ x &= \left[\sum_{j} CO2\_1(j) + \sum_{k} CO2\_2(k) + \sum_{l} CO2\_3(l)\right] / \\ \left[\sum_{j} Flow\_gas(j) + \sum_{k} Flow\_coal(k) + \sum_{l} Flow\_ind(l)\right] \\ y &= \left[\sum_{j} CO2CAPEX1(j) + \sum_{k} CO2CAPEX2(k) + \sum_{l} CO2CAPEX3(l)\right] / \\ \left[\sum_{j} Flow\_gas(j) + \sum_{k} Flow\_coal(k) + \sum_{l} Flow\_ind(l)\right] \\ \end{split}$$
(10)

$$z = \left[\sum_{j} CO2OPEX1(j) + \sum_{k} CO2OPEX2(k) + \sum_{l} CO2OPEX3(l)\right] / \left[\sum_{j} Flow_{gas}(j) + \sum_{k} Flow_{coal}(k) + \sum_{l} Flow_{ind}(l)\right]$$
(11)

[Table 9 about here.]

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## List of Figures

1	Illustrative diagram of the oxy-combustion capture process highlighting the main energy intensive units and revenue sources and outputs for each island: the ASU island and the $CO_2CPU$	97
2	Flow diagram of a compression and dehydration process con- sisting of a three-stage pre-compression train, a dehydration unit and a final product compression train applied to both a post combustion product stream and an oxy-combustion de-	37
3	Bar chart comparing installed equipment cost and electricity costs of the oxy-combustion and the post combustion 6-stage	38
4	compression and dehydration CPU systems $\ldots$ $\ldots$ $\ldots$ Flow diagram of a double flash CO <sub>2</sub> CPU system model with heat integration consisting of a 3 stage pro compression train	39
	a double flash separation system and dehydrator and a fi- nal product compression train resulting in a high purity $CO_2$	
5	product stream $\ldots$	40
6	train	41
0	Distillation Column separation process using heat integration, a dehydrator and pre & post processing compression	42
7	Graph showing the $CO_2$ price per tonne sold if marketed for EOR assuming a minimum rate of return on investment of 20%, 15%, 10% and 5% on the $CO_2CPU$ as a function of $CO_2$	
8	stream purity	43
	system	44
9	Illustration of a UK based $CO_2$ transport network scenario [56]	45
10	Graph Showing Network Capture System CAPEX and OPEX	
	per tonne of $OO_2$ Captured as a Function of Minimum Trunk	16
	Line Furity - Pareto Front	40

11	Graph showing the percentage reduction in $CO_2$ price when
	choosing a minimum cost scenario as opposed to a maximum
	purity scenario for Internal Rates of Return of 20%, 15%, 10%
	and $5\%$



Figure 1: Illustrative diagram of the oxy-combustion capture process highlighting the main energy intensive units and revenue sources and outputs for each island: the ASU island and the  $CO_2CPU$  island.



Figure 2: Flow diagram of a compression and dehydration process consisting of a threestage pre-compression train, a dehydration unit and a final product compression train applied to both a post combustion product stream and an oxy-combustion derived flue gas stream



Figure 3: Bar chart comparing installed equipment cost and electricity costs of the oxycombustion and the post combustion 6-stage compression and dehydration CPU systems



Figure 4: Flow diagram of a double flash  $CO_2CPU$  system model with heat integration consisting of a 3-stage pre compression train, a double flash separation system and dehydrator and a final product compression train resulting in a high purity  $CO_2$  product stream



Figure 5: Flow diagram of a double flash  $CO_2CPU$  system model without heat integration producing a low purity  $CO_2$  product, consisting of a 3-stage pre-compression train, a double flash separation system and dehydrator and a final product compression train



Figure 6: Flow Diagram of a  $CO_2CPU$  system model with a 6-Stage Distillation Column separation process using heat integration, a dehydrator and pre & post processing compression



Figure 7: Graph showing the  $CO_2$  price per tonne sold if marketed for EOR assuming a minimum rate of return on investment of 20%, 15%, 10% and 5% on the  $CO_2CPU$  as a function of  $CO_2$  stream purity



Figure 8: Flow diagram of the triethylene dehydration system used to dehydrate pressurized flue gas, consisting of an absorber-stripper system



Figure 9: Illustration of a UK based  $CO_2$  transport network scenario [56]



Figure 10: Graph Showing Network Capture System CAPEX and OPEX per tonne of CO<sub>2</sub> Captured as a Function of Minimum Trunk Line Purity - Pareto Front



Figure 11: Graph showing the percentage reduction in  $CO_2$  price when choosing a minimum cost scenario as opposed to a maximum purity scenario for Internal Rates of Return of 20%, 15%, 10% and 5%

## List of Tables

1	Typical oxy-combustion flue gas composition from a coal fired	
	power plant (a.), typical $CO_2$ composition for EOR (b.) and	
	Dynamis $CO_2$ stream quality recommendations (c.)	49
2	Example of flue gas properties from an oxy-combustion pulver-	
	ised coal firing power plant at nominal load generating 347MWe.	50
3	$CO_2$ product stream characteristics from amine-based post	
	combustion capture process $[32]$	51
4	System Performance and economic evaluation of all $CO_2CPUs$	
	modeled (*separation efficiency only applies to the $CO_2CPU$	
	of the post combustion system)	52
5	Power plant net efficiency losses and $CO_2$ separation efficien-	
	cies resulting from all of the $CO_2CPUs$ modeled (*separation	
	efficiency only applies to the $CO_2CPU$ of the post combustion	
	system)	53
6	Capital and Operational costs of the Triethylene Glycol De-	
	hydration system compared with overall costs for each eco-	
	nomically worthwhile CPU process (excluding the low purity	
	double flash CPU scenario)	54
7	UK Case Study: Flow Rates of Gas CCGT Plants, Coal Plants	
	and Steel Plant	55
8	Aspen HYSYS Process Utilities	56
9	Optimization Variables for Transport Network Problem	57

	c.Dynamis recommendations for CO <sub>2</sub> quality by volume [28]		>0.955	<4%	<4% (saline aquifers) 100-1000 ppm (EOR)	$<\!4\%$	<100 ppm	<500 ppm
	b.Weyburn EOR project CO <sub>2</sub> composition by volume [54]		>0.960	<300 ppm	<50~ m ppm			$< 20 \mathrm{~ppm}$
quality recommendations (c.)	a.Flue gas composition range by mass (volume) from coal fired	oxy-combustion plants [28]	0.793- $0.824(0.724$ - $0.764)$	0.078 - 0.099 (0.113-0.142)	0.061-0.076 (0.078 - 0.095)	0.028 - 0.031 (0.028 - 0.031)	$\begin{array}{c} 200 \text{ ppm} \\ (100 \text{ ppm}) \end{array}$	0.048 - 0.059 ( $0.010 - 0.014$ )
(b.) and Dynamis CO <sub>2</sub> stream			CO2	$\mathrm{N}_2$	$O_2$	Ar	$\mathrm{SO}_2$	$\mathrm{H}_{2}O$

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Table 2: Example of flue gas properties from an oxy-combustion pulverised coal firing power plant at nominal load generating 347MWe.

Flue Gas Porperty (CPU inlet)	
Temperature (Celsius)	13.2
Pressure (bar)	1
Flow Rate (tonne/hour)	342.7
Composition (mass fraction/mole fraction)	
$\mathrm{CO}_2$	0.824/0.764
$O_2$	0.061/0.078
$N_2$	0.080/0.113
Ar	0.031/0.031
$H_2$	0.006/0.013
$SO_2$	$200({ m ppm})/100~({ m ppm})$

Table 3:  $CO_2$  product stream characteristics from amine-based post combustion captureprocess [32] $\boxed{CO_2(1,27) = W_2(1,27) = W_2(1,27) = O_2(1,27) = O_2(1,$ 

$CO_2$ (wt%)	H <sub>2</sub> O (wt%)	N <sub>2</sub> (wt%)	$O_2$ (wt%)	Temperature	Pressure
	1120 (11070)	1.2 (	02(	(°C)	(bar)
98.5	1.5	200  ppm	100  ppm	35	2

Table 4: System Performat	nce and economic evalu	iation of all CO <sub>2</sub>	CPUs modeled	(*separation efficiency	only applies to the
CO <sub>2</sub> CPU of the post combi	ustion system)			,	
Property of CPU	<b>Distillation</b> CPU	Double Fl	ash CPU	Compression and	Dehydration Unit
		High Purity	Low Purity	Oxy-Combustion	Post Combustion
CO <sub>2</sub> product stream Mt/vear	2.16	2.25	2.49	2.88	2.35
Separation <sup>É</sup> fficiency	30%	92%	97%	100~%	$100\%^*$
$CO_2$ Purity	99.98 wt.%	97.54  wt.%	92.55  wt.%	82.91 wt.%	99.97  wt.%
Oxygen Content	$0.4 \mathrm{~ppm}$	1.03  wt.%	2.89  wt.%	6.18  wt.%	150  ppm
Total Capital Cost	${ m E51M}$	${ m f34M}$	f42M	${ m f33M}$	f23M
Total Yearly Operating Cost	f34M	$\mathrm{f}32\mathrm{M}$	f36M	f28M	£17M

-dae ) nati		and Dehydration Unit	1 Post Combustion	100%*	99.97  wt.%	96*	3% CPU	6% Regeneration 1% Solvent Transport
		Compression	Oxy-Combustion	100~%	82.91  wt.%	103	4.5% CPU	5% ASU [22]
suttang rrom an u n)		rlash CPU	Low Purity	97%	92.55  wt.%	158	6.0% CPU	5% ASU [22]
rombitstion system		Double F	High Purity	92%	97.54  wt.%	150	5.1% CPU	5% ASU [22]
Disses ally UO2 separate P.C.O.C.P.I. of the nost		Distillation CPU		90%	99.98 wt.%	172	5.7% CPU	5% ASU [22]
able 3. I Ower prain life entitiently aration efficiency only applies to the	with on gotted day fitter fortester the man	Property of CPU		Separation Efficiency	CO <sub>2</sub> Purity	Unit Energy (kWh/tCO <sub>2</sub> captured yearly)	Power Plant Net Efficiency Loss Breakdown	

 Table 5: Power plant net efficiency losses and CO<sub>2</sub> separation efficiencies resulting from all of the CO<sub>2</sub>CPUs modeled (\*separation efficiency only applies to the CO<sub>2</sub>CPU of the post combustion system)

Table 6: Capital and Operational costeconomically worthwhile CPU process	s of the Triethylene Glyc (excluding the low purity	ol Dehydration system c double flash CPU scenar	ompared with overall costs for io)	: each
	TEG Dehydration	% of total CD CPU	% of total HPDF CPU	% of total Dist CPU
Yearly Operating Costs	£178,498	0.6%	0.6%	0.5%
Total Installed Equipment Cost	f(727,900)	2.2%	2.1%	1.4%

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e 6: Capital and Operational costs of the Triethylene Glycol Dehydration system compared $\mathbf{w}$	omically worthwhile CPU process (excluding the low purity double flash CPU scenario)
abl	con

Gas Plant (J)	Flow rate $(MtCO_2/yr.)$
Keadby	1.5
Didcot B	2.6
South Humber Bank	2.6
Barking	2.4
Killingholme A	1.3
Sutton Bridge	1.8
Damhead Creek	1.7
$\operatorname{Spalding}$	1.7
Coryton	1.5
Little Barford	1.3
Coal Plant (K)	Flow rate $(MtCO_2/yr.)$
West Burton	7.8
Cottam	8.4
Drax	19.4
m Kingsnorth	6.3
Ratcliffe	7.7
Rugeley	3.8
DidcotA	4.7
Ferrybridge C	6
Eggborough	6.5
Tilbury	3.8
Steel Plant (L)	Flow rate $(MtCO_2/yr.)$
Corus	5.8

Table 7: UK Case Study: Flow Rates of Gas CCGT Plants, Coal Plants and Steel Plant.

Table 8: Aspen HYSYS Process Utilities				
Name	Fluid Type	Conditions Pressure,Temperature	Cost	Units
Electricity			0.0576	$\pounds/kW$
Cooling Water	Water	$105 \mathrm{kPa}_g; 20^{\circ}\mathrm{C}$	0.00443	$\pounds/tonne$
Propane	$\operatorname{Refrigerant}$	$105 \mathrm{kPa}_g;$ -40°C	0.0580	$\pounds/tonne$
LP Steam	Steam	$0 \mathrm{kPa}_g; 125 \ \mathrm{^{\circ}C}$	0.002	$\pounds/MJ$
Refrigerant 1	Propane	$0 \text{ kPa}_g; -24^{\circ}\text{C}$	0.003	$\pounds/MJ$
Ethane	$\operatorname{Refrigerant}$	$105 \text{ kPa}_{g}; -90^{\circ}\text{C}$	0.036000	$\pounds/tonne$
Freon 12	Refrigerant	$105 \text{ kPa}_{g}; -29.8^{\circ}\text{C}$	0.170000	$\pounds/tonne$

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 Table 9: Optimization Variables for Transport Network Problem.

Optimization	Description
Variable	
$CO2_1(J)$	Purity of $CO_2$ stream from sum of gas plants (J) with post
	combustion capture (V) in mass fraction
$CO2_1(K)$	Purity of CO <sub>2</sub> stream from sum of coal plants (K) with oxy-
	combustion capture (U) in mass fraction
$\rm CO2\_1(L)$	Purity of $CO_2$ stream from industrial plants (L) with oxy-
	combustion capture (U) in mass fraction
CO2Capex1 (J)	Total CAPEX from gas plants (J) using technology V in $\pounds M$
	per $Mt_{CO_2}$ captured per year
CO2Capex2 (K)	Total CAPEX from coal plants (K) using technology U in $\pounds M$
	per $Mt_{CO_2}$ captured per year
CO2Capex3 (L)	Total CAPEX from industrial plants (L) using technology U
	in $\pounds M$ per $Mt_{CO_2}$ captured per year
CO2Opex1 (J)	Total OPEX from gas plants $(J)$ using technology V in £M
	per $Mt_{CO_2}$ captured per year
CO2Opex2 (K)	Total OPEX from coal plants (K) using technology U in $\pounds M$
	per $Mt_{CO_2}$ captured per year
CO2Opex3 (L)	Total OPEX from industrial plants (L) using technology U in
	$\pounds M$ per $Mt_{CO_2}$ captured per year